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TUNABLE LASERS

Final Report

for the period 1 July 1974 - 30 June 1976

by

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The objectives of this research program were to develop wavelength-tunable laser systems and to explore their applications. During the program, we completed our investigation of $KB_5O_8 \cdot 4H_2O$ and $NH_4B_5O_8 \cdot 4H_2O$ as phase-matchable nonlinear crystals for frequency doubling to 217 nm. We demonstrated further the utility and potential of rotationally-twinned $\bar{4}3$ m nonlinear crystals by frequency-doubling a tunable dye laser in ZnSe. An enhancement of 1500 above that obtained in a single crystal was obtained.

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Further calculations on optimum crystallographic orientatins for nonlinear effects in twinned crystals were completed. A new method of obtaining 4 regularly-twinned domains in a single crystal element was devised. These small elements may be optically contacted to produce the effect of 4, 8, 16, ... regular domains. Proof was obtained that twinned seed crystals can be grown to larger dimensions. Experiments were conducted which proved that no damage or anomalous absorption occurs at twins. Work continued on the two photon creation of carriers in ZnSe. It was found that this carrier production method which is dominant at high power levels is very sensitive to wavelength in the 9000 Å region, falling off rapidly to about 1/10th the value at 9200 Å that it had at 9000 Å. These measurements imply that the saturation in infrared difference frequency generation, which we believe to be due to absorption by these free carriers, can be substantially reduced by using longer wavelength pump lasers. Research was conducted on infrared laser dyes and bleaching probabilities and efficiencies were determined for 16 promising compounds. We succeeded in producing continuously-tunable infrared radiation from 11 μm to 21 μm in proustite with useful efficiency. Finally, a review and extension of previous work on optoacoustic spectroscopy was completed.

TABLE OF CONTENTS

I. INTRODUCTION

Our work over the past several years has emphasized the production and detection of powerful wavelength-tunable laser sources. The practical applications of these techniques are varied, and include:

- Laser-induced chemical reactions using wavelength-tunable lasers
- Isotope separation and other processes requiring high-power laser sources at selected wavelengths
- High-resolution infrared spectroscopy
- Studies of surface catalysis processes
- Multiple-channel optical communication systems
- Detection of atmospheric pollutants
- Quantitation of laser absorption by atmospheric constituents

II. RESEARCH ON ROTATIONALLY-TWINNED CRYSTALS

The basic physical process which we have employed for infrared generation is to produce the difference-frequency between two laser sources in an appropriate nonlinear optical medium. An important aspect of the difference-frequency method is that it is inherently efficient; in principle,

100% of the photons of the pump laser can be converted to photons at the difference frequency. Practical realization of this advantage is inhibited by the nature of the nonlinear optical materials in which mixing may be accomplished.

Conventional phase-matchable nonlinear optical crystals suitable for phase-matched difference-frequency generation have several practical difficulties. Most have relatively modest nonlinear coefficients and high refractive indices, so that the figure of merit, (d^2/n^3), is low. Increases in the optical power density of the mixing beams is often not possible because of the low damage threshold of the crystals. Also, the absorption coefficient of such crystals is frequently large, up to several cm^{-1} in some cases. And, finally, the crystals are generally difficult to grow and expensive. The above facts suggest that progress in the development of suitable conventional phase-matching crystals is possible and worthy of continued support; but alternative approaches should also be vigorously explored.

The results of our research on rotationally-twinned $\bar{4}3m$ crystals are reported in Refs. 1, 3, and 4 cited in Appendix A. A simple theory was developed which accounts for the induced polarization of different domains in a rotationally-twinned crystal. The experimental results obtained prior to the beginning of this contract in

difference-frequency generation between a ruby laser and a dye laser in ZnSe were correlated with the theory and published in Ref. 1. A second confirmatory experiment in ZnSe, frequency doubling a dye laser tunable from 0.88 μm - 1.04 μm, was correlated with a simple theory which includes the effects of crystal absorption. These results appear in Ref. 3.

Further work on classifying suitable $\bar{4}3m$ crystals exhibiting rotational twins was completed. These results are summarized in Ref. 5. Growth methods for replicating crystals having favorable rotational twin spacings were investigated and evidence was obtained that such replication can be accomplished. This finding confirms that rotationally-twinned nonlinear optical crystals may be fabricated in a scalable way. A useful method of producing crystal elements containing the equivalent of at least 4 coherent domains was developed. Experimental verification of this concept was not possible during the contract, but additional work is contemplated.

We have specifically examined the power handling capabilities and potential damage mechanisms of rotationally-twinned crystals. ZnSe samples were tested to better than 100 mW/cm^2 power levels (the limit imposed by surface damage of our specimens) with no internal effects associated with the rotational twins. Theoretical considerations also

suggest no deleterious effects up to the power levels associated with bulk damage (~ 1 GW/cm^2).

Effects of bulk absorption by second harmonic near the crystal band gap were studied and reported in Technical Report #2 for the period ending June 30, 1975. Crystal electrical conductivity was increased by the two-photon production of free carriers if the two-photon energy was below the band gap of the material. This explains the saturation of difference-frequency radiation we observed with high incident power levels in Ref. 1. By using pump lasers at longer wavelengths, higher conversion efficiencies could be obtained.

A U. S. Patent on the use of rotationally-twinned crystals to produce nonlinear optical effects was recently granted to the Principal Investigator (Ref. 9).

III. OPTOACOUSTIC SPECTROSCOPY

Refs. 6, 7, 8, and 10, of Appendix A report further results on optoacoustic spectroscopy. This measurement technique has been found useful in measuring the atmospheric absorption of high-power laser beams, in diagnostic experiments, in stabilizing laser lines, and in detecting atmospheric pollutants. One patent was granted (Ref. 10). An additional patent on acoustically-resonant spectrophones was filed (Ref. 6).

Theoretical investigation of the mechanisms for resonant signal enhancement are reported in Refs. 7 and 8. The sources of loss and origins of the observed background signals were correlated with the data obtained in our laboratory and elsewhere. A particularly useful method of overcoming background signals caused by both window absorption and the wings of strong nearby lines is described in Ref. 8 and is the subject of a pending U. S. Patent (Ref. 6). The method is based on wide modulation of the source wavelength, producing an acoustic signal at twice the frequency of modulation if the absorption line of interest is centered within the bandwidth of modulation. A future experimental test of this concept is contemplated.

IV. CATEGORIZATION OF NEAR-INFRARED LASER DYES

The lasing efficiencies and photochemical stabilities of laser dyes useful in the 710-1080 nm spectral region were investigated using a ruby laser as the optical pump. The measured bleaching rates (defined as the probability of irreversible decomposition of a dye molecule per absorbed photon) varied from $\leq 1 \times 10^{-5}$ to 3×10^{-4} for the different dye-solvent combinations investigated. Broad band lasing efficiencies ranged from 4 to 43%. Shifts of wavelength tuning range with variations in solvent, dye

concentration, and dye laser cavity geometry were investigated. These results are reported in Ref. 2.

Previous to our investigation, laser dyes in the near infrared region of the spectrum we believed to be highly unstable and subject to rapid photochemical bleaching. We have demonstrated that some selected dyes spanning the 710-1000 nm spectral region exhibit bleaching rates as low as many dyes used satisfactorily in the visible.

V. NONLINEAR OPTICAL MIXING IN PROUSTITE

A conventional view of nonlinear optical materials suggests that they are useful only in the spectral region between the band gap and the shortest wavelength multiphonon absorption band. We recognized, however, that many of these crystals, such as proustite, have useful transparency at wavelengths beyond the first absorption band. For this reason, we undertook to demonstrate that useful nonlinear optical power may be generated in crystals at these longer wavelengths. Our results are reported in Ref. 5.

Continuously tunable infrared radiation was produced in the spectral range 11 μm to 23 μm by nonlinear mixing of two ruby-pumped dye lasers in proustite. A peak power of about 3 watts was observed at 19 μm . A 7^{o} crystal angle

change was sufficient to achieve phase matching over the entire spectral region. A theory including the angular divergence of the mixing beams and the absorption of the crystal was developed, and excellent correlation was obtained between the theory and the experimental results.

Appendix A. List of publications resulting from research under this contract

1. C. F. Dewey, Jr. and L. O. Hocker, "Enhanced nonlinear optical effects in rotationally-twinned crystals," Appl. Phys. Lett., 26, 442 (1975).

2. P. E. Oettinger and C. F. Dewey, Jr., "Lasing efficiency and photochemical stability of infrared laser dyes in the 710-1080 nm spectral region," IEEE J. Quant. Elect., QE-12, 95 (1976).

3. L. O. Hocker and C. F. Dewey, Jr., "Enhancement of second-harmonic generation in zinc selenide by crystal defects," Appl. Phys. Lett., 28, 267 (1976).

4. C. F. Dewey, Jr., "Nonlinear optical effects in rotationally-twinned crystals: an evaluation of CdTe, ZnTe, and ZnSe," J. de Physique: Revue de Physique Appliquee, 12 (to appear) (1977).

5. L. O. Hocker and C. F. Dewey, Jr., "Difference-frequency generation from 11 μm to 23 μm in proustite," Appl. Phys., 11, 137 (1976).

6. C. F. Dewey, Jr. and L. O. Hocker, Optoacoustic spectroscopy employing amplitude and wavelength modulation, U. S. Patent pending.

7. R. D. Kamm, "Detection of weakly absorbing gases using a resonant optoacoustic method," J. Appl. Phys., 47, 3550 (1976).

8. C. F. Dewey, Jr., "Design of Optoacoustic Systems", Chapt. 4, in Optoacoustic Spectroscopy, edit. by Y.-H. Pao, Academic Press, N. Y., Fall 1977.

9. C. F. Dewey, Jr., Nonlinear Amplifying, U. S. Patent 3,988,593 (October 26, 1976).

10. C. F. Dewey, Jr., Resonant Opto-Acoustic Detection System, U. S. Patent 3,938,365 (17 February 1976).